Stratospheric Ozone Variations Caused by Solar Proton Events Between 1963 and 2005

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Abstract Some solar eruptions lead to solar proton events (SPEs) at the Earth, which typically last a few days. High energy solar protons associated with SPEs precipitate on the Earth's atmosphere and cause increases in odd hydrogen (HO_x) and odd nitrogen (NO_y) in the polar cap regions (>60° geomagnetic). The enhanced HO_x leads to short-lived ozone depletion (~days) due to the short lifetime of HO_x constituents. The enhanced NO_y leads to long-lived ozone changes because of the long lifetime of the NO_y family in the stratosphere and lower mesosphere. Very large SPEs occurred in 1972, 1989, 2000, 2001, and 2003 and were predicted to cause maximum total ozone depletions of 1–3%, which lasted for several months to years past the events. A long-term data set of solar proton fluxes used in these computations has been compiled for the time period 1963–2005. Several satellites, including the NASA Interplanetary Monitoring Platforms (1963–1993) and the NOAA Geostationary Operational Environmental Satellites (1994–2005), have been used to compile this data set.

1 Introduction

Explosions on the Sun sometimes result in large fluxes of high-energy solar protons at the Earth, especially near the maximum period of activity of a solar cycle. This disturbed time, wherein the solar proton flux is generally elevated for a few days, is known as a solar proton event (SPE). Solar protons are guided by the Earth's magnetic field and impact both the northern and southern polar cap regions (>60° geomagnetic latitude) (e.g., see Jackman and McPeters 2004). These protons can impact the neutral middle atmosphere (stratosphere and mesosphere) and produce

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both HO_x (H, OH, HO_2) and NO_y (N, NO, NO_2, NO_3, N_2O_5, HNO_3, HO_2NO_2, $ClONO_2$, $BrONO_2$) constituents either directly or through a photochemical sequence (e.g., Swider and Keneshea 1973; Crutzen et al. 1975; Jackman et al. 1980; Solomon et al. 1981; McPeters 1986; Zadorozhny et al. 1992). Ozone is also impacted by the solar protons through direct photochemical destruction forced by the HO_x and NO_y enhancements (e.g., Weeks et al. 1972; Heath et al. 1977; Solomon et al. 1983; Jackman et al. 1990).

Although all sizes of SPEs can have an impact on the atmosphere, the extremely large SPEs cause the most pronounced changes. Several of these extremely large SPEs have occurred in the past 40 years. Huge fluxes of high-energy protons have impacted the Earth's atmosphere in 1972, 1989, 2000, 2001, and 2003. In this paper, the impact of SPEs over the 1963–2005 period will be discussed, concentrating particularly on the atmospheric effects during and after the huge SPEs.

The paper is divided into six primary sections, including the Introduction. We discuss the very important solar proton measurements and their production of odd hydrogen (HO_x) and odd nitrogen (NO_y) in Sect. 2. A comparison of the largest 15 SPEs in the past four solar cycles is also undertaken in Sect. 2. The GSFC two-dimensional model used to simulate the impact of the SPEs on the atmosphere is discussed in Sect. 3. The short-term impact of these SPEs on ozone during and for several days after particular events is given in Sect. 4. Longer term influences of the SPEs on the middle atmosphere are discussed in Sect. 5. Finally, the conclusions are given in Sect. 6.

2 Proton Fluxes: Odd Hydrogen (HO_x) and Odd Nitrogen (NO_y) Production

2.1 *Proton Fluxes*

Solar proton fluxes have been measured by a number of satellites in interplanetary space or in orbit around the Earth. The National Aeronautics and Space Administration (NASA) Interplanetary Monitoring Platform (IMP) series of satellites provided measurements of proton fluxes from 1963–1993. IMPs 1–7 were used for the fluxes from 1963–1973 (Jackman et al. 1990) and IMP 8 was used for the fluxes from 1974–1993 (Vitt and Jackman 1996). The National Oceanic and Atmospheric Administration (NOAA) Geostationary Operational Environmental Satellites (GOES) were used for proton fluxes from 1994–2005 (Jackman et al. 2005a). There are uncertainties associated with these proton flux data, especially given the large number of satellite instruments used to compile such a measurement record. We estimate the proton flux uncertainties to be up to 50%, given some straightforward comparisons of particular proton flux measuring instruments. It is beyond the scope of the present study to undertake a more detailed comparison, however, we do recommend that such a study be accomplished by experts in the field of solar particle observations.

Other precipitating particles are associated with SPEs, besides protons. Alpha particles comprise about 10% of the positively charged solar particles with other ions accounting for less than 1% of the remainder (e.g., Mewaldt et al. 2005). Measurements exist for other ions besides protons, but not enough for a continuous observation record from 1963–2005. Electrons also accompany SPEs (e.g., Mewaldt et al. 2005), however, their precipitation is not uniform over the polar caps. The energy deposition of electrons will primarily be in the auroral oval regions, which is more difficult to characterize, and will be confined mostly to the thermosphere and upper mesosphere. We only include solar protons in our computations and note that other charged particles could add modestly to the energy deposition in the middle atmosphere during SPEs.

Protons in their energy deposition process cause ionizations, dissociations, predissociations, and dissociative ionizations in collisions with atmospheric constituents. The protons thus produce secondary electrons, ions, excited molecules and atoms. The proton fluxes were used to compute daily average ion pair production profiles using an energy deposition scheme first discussed in Jackman et al. (1980). The scheme includes the deposition of energy by the protons and assumes 35 eV are required to produce one ion pair (Porter et al. 1976). Thereby, a data set of daily average ion pair production rates for the period 1963–2005 were created for use in model studies and is available at http://strat-www.met.fu-berlin.de/~matthes/sparc/inputdata.html.

2.2 *Odd Hydrogen (HO_x) Production*

Along with the ion pairs, the protons and their associated secondary electrons also produce odd hydrogen (HO_x). The production of HO_x relies on complicated ion chemistry that takes place after the initial formation of ion pairs (Swider and Keneshea 1973; Frederick 1976; Solomon et al. 1981). Solomon et al. (1981) computed HO_x production rates as a function of altitude and ion pair production. Each ion pair typically results in the production of around two HO_x constituents in the upper stratosphere and lower mesosphere. In the middle and upper mesosphere, an ion pair is computed to produce less than two HO_x constituents per ion pair. We include the HO_x production by SPEs in our model using a look-up table (see Jackman et al. 2005b) invoking the computations of Solomon et al. (1981). The HO_x constituents have lifetimes of only hours in the middle atmosphere, therefore, any further effects on other constituents from the HO_x group are apparent only during and shortly after an SPE.

2.3 *Odd Nitrogen (NO_y) Production*

Odd nitrogen is produced when the energetic charged particles (protons and associated secondary electrons) collide with and dissociate N_2. Following Porter et al. (1976) it is assumed that ~1.25 N atoms are produced per ion pair. The Porter et al. (1976) study also further divided the proton impact of N atom production between ground state (~45% or ~0.55 per ion pair) and excited state (~55% or ~0.7 per ion pair) nitrogen atoms. Ground state [$N(^4S)$] nitrogen atoms can create other NO_y constituents, such as NO, through

$$N(^4S) + O_2 \rightarrow NO + O \tag{1}$$

or can lead to NO_y destruction through

$$N(^4S) + NO \rightarrow N_2 + O. \tag{2}$$

Generally, excited states of atomic nitrogen, such as $N(^2D)$, result in the production of NO through

$$N(^2D) + O_2 \rightarrow NO + O \tag{3}$$

(e.g., Rusch et al. 1981; Rees 1989) and do not cause significant destruction of NO_y. Rusch et al. (1981) showed that there are huge differences in the final results of model computations of NO_y enhancements from SPEs that depend strongly on the branching ratios of the N atoms produced. We currently do not include any of the excited states of atomic nitrogen (e.g., $N(^2D)$, $N(^2P)$, and N^+) as computed constituents in our model. We use the following fairly accurate way to best represent the production of NO_y constituents by the SPEs: Assume that 45% of the N atoms produced per ion pair result in the production of $N(^4S)$ (~0.55 per ion pair) and that 55% of the N atoms produced per ion pair result in the production of NO (~0.7 per ion pair). There are uncertainties in these proton-caused NO_y production computations of about 20% (Jackman et al. 1979), which does not include the uncertainty in the measured precipitating proton flux.

The lifetime of odd nitrogen can vary dramatically depending on season and altitude. Odd nitrogen has a relatively short lifetime (~days) in the sunlit middle and upper mesosphere, however, lower mesospheric and stratospheric NO_y can last for weeks past an SPE. A large portion of the SPE-produced NO_y is conserved in a mostly dark polar middle atmosphere in the late fall and winter. This NO_y can then be transported to lower altitudes via the general downward flowing winds during this time of year and its lifetime can range from months to years, if transported all the way to the middle and lower stratosphere.

We have quantified middle atmospheric NO_y production previously (Jackman et al. 1990; Vitt and Jackman 1996; Jackman et al. 2005a) for years 1963 through 2003. We add NO_y computations in this study to these earlier calculations for years 2004 and 2005 and present the annual production from SPEs for the 43-year period 1963 through 2005 in Fig. 1. The annual-averaged sunspot number is also shown in Fig. 1 to illustrate the rough correlation between solar maximum periods and frequency of SPEs.

Substantial amounts of NO_y were produced near solar maximum in several years. The annual global NO_y production from solar protons is computed to be 3.7, 8.4, 6.7, 7.9, and 4.1 × 10^{33} molecules for the very active years 1972, 1989, 2000, 2001, and 2003, respectively. These annual production rates from SPEs are ~10–25%

of the largest global NO_y source (nitrous oxide oxidation, $N_2O + O(^1D)$) of about 3.3×10^{34} molecules/year (Vitt and Jackman 1996). The SPE sources of NO_y were very significant during these particular years for the middle atmosphere. Since the SPEs typically last only a few days, these impulses of NO_y from SPEs can impact the polar odd nitrogen amounts substantially during brief periods.

The 15 largest SPEs based on NO_y production in the past 40 years are given in Table 1. Surprisingly, eight of them occurred in the most recent solar maximum period.

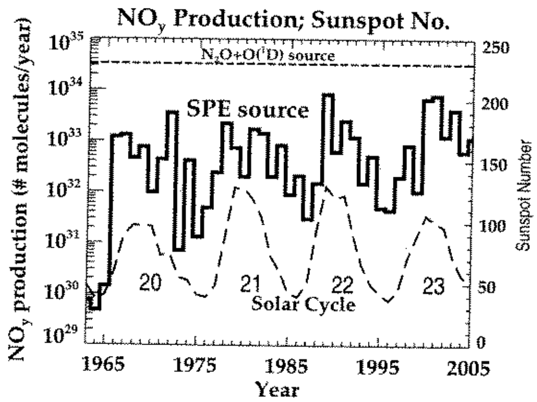

Fig. 1 Total global production of NO_y molecules per year in the polar stratosphere and mesosphere by SPEs (solid histogram – left ordinate) and the oxidation of nitrous oxide $[N_2O + O(^1D)]$ (dash-dot line – left ordinate) for years 1963–2005. The annually averaged sunspot number (dashed line – right ordinate) is also given.

Table 1 Largest 15 solar proton events in the past 40 years

Date of SPEs	Rank in size	NO_y production in the middle atmosphere (# of 10^{33} molecules)
October 19–27, 1989	1	6.7
August 2–10, 1972	2	3.6
July 14–16, 2000	3	3.5
October 28–31, 2003	4	3.4
November 5–7, 2001	5	3.2
November 9–11, 2000	6	2.3
September 24–30, 2001	7	2.0
August 13–26, 1989	8	1.8
November 23–25, 2001	9	1.7
September 2–7, 1966	10	1.2
January 15–23, 2005	11	1.1
September 29–October 3, 1989	12	1.0
January 28–February 1, 1967	13	0.99
March 23–29, 1991	14	0.89
September 7–17, 2005	15	0.88

3 GSFC Two-Dimensional Model Description and Simulations

The latest version of the Goddard Space Flight Center (GSFC) two-dimensional (2D) atmospheric model was used to predict atmospheric changes caused by the solar protons. The model has been in use since the late 1980s and has undergone extensive improvements over the years (Douglass et al. 1989; Jackman et al. 1990). The vertical range of the model, equally spaced in log pressure, is from the ground to approximately 90 km (0.0024 hPa) with approximately a 2 km grid spacing. Latitudes range from 85°S to 85°N with a 10°-grid spacing.

Fleming et al. (2002) described the methodology to compute the transport for the GSFC 2D model using the global winds and temperatures from meteorological data for particular years. This technique has now been applied using the National Centers for Environmental Prediction – National Center for Atmospheric Research (NCEP-NCAR) reanalysis-2 project (e.g., Kanamitsu et al. 2002). These data cover the time period 1958–present, and extend from the surface to 10 hPa. We have used the original NCEP analyses data (Gelman et al. 1986) for 10–1 hPa for 1979–present (climatological fields are used above 10 hPa prior to 1979). For the mesosphere for 1–0.002 hPa, we employ the temperature measurements made by the Microwave Limb Sounder (MLS) onboard UARS for September 1991 through June 1997 (Wu et al. 2003). The 2D model residual circulation and horizontal and vertical eddy diffusion quantities are then derived following the methodology described in Fleming et al. (2002, 2007).

The photochemical scheme includes all reactions that are thought to be important for ozone in the middle atmosphere. The reaction rates, including heterogeneous rates, are taken from Sander et al. (2003). A lookup table is employed in computing the photolytic source term, which is then used in computation of photodissociation rates for atmospheric constituents (Jackman et al. 1996). The GSFC 2D chemistry solver uses the Atmospheric Environmental Research (AER) 2D model scheme (Weisenstein et al. 2004), which computes a diurnal cycle every day. The ground boundary conditions for the source gases are taken from WMO (2003) for the particular simulated year. The model uses chemical families and computes 62 constituents (Jackman et al. 2005b).

We used the GSFC 2D model to compute two primary simulations, "base" and "perturbed," for the years 1960–2010. The transport for years 1960–2004 is driven by NCEP products for those particular years, whereas the transport for the individual years 2005–2010 is an average climatology of the 1958–2004 period. The "base" simulation includes no SPEs, whereas the "perturbed" simulation includes all SPEs from January 1, 1963 through December 31, 2005. The perturbation to the atmosphere was caused by the SPE-produced HO_x and NO_y enhancement.

4 Short-term Impact on Ozone

The ozone response due to very large SPEs is not subtle and has been observed due to numerous events to date (e.g., Jackman and McPeters 2004; López-Puertas et al. 2005; Seppälä et al. 2006). Ozone within the polar caps (60–90°S or 60–90°N geomagnetic) is generally depleted to some extent in the mesosphere and upper stratosphere (e.g., Jackman et al. 2005b) within hours of the start of the SPE. Decreases in mesospheric and upper stratospheric ozone are mostly caused by SPE-induced HO_x increases (see Solomon et al. 1981, 1983; Jackman and McPeters 1985; Jackman et al. 2005b; Verronen et al. 2006) and last only during and for a few hours after the SPEs. The Verronen et al. (2006) study implies that the solar proton-caused HO_x production could be up to 50% uncertain, which does include the uncertainty in the measured precipitating proton flux. SPE-caused NO_y enhancements can also drive upper stratospheric ozone depletion, but do not cause significant mesospheric ozone depletion (Jackman et al. 2001). Although these short-term SPE impacts on ozone merit study and have helped test atmospheric models (e.g., Jackman and McPeters 1987; Verronen et al. 2006), the longer-term SPE impacts on ozone are the more important component in polar stratospheric ozone variation and will be discussed in the next section.

5 Long-term Impact on Ozone

The longer-term impact of SPE-induced NO_y enhancements on ozone has been known for about 30 years. Heath et al. (1977) showed large stratospheric ozone reductions in Nimbus-4 BUV instrument data up to 19 days past the August 1972 events, which were probably caused by the NO_y enhancements. Several other papers (e.g., Reagan et al. 1981; Solomon and Crutzen 1981; Rusch et al. 1981; Jackman et al. 1990, 1995, 2000, 2005a; Randall et al. 2001) studied various aspects of NO_y influence on stratospheric ozone. The primary catalytic cycle for NO_y destruction of ozone is

$$NO + O_3 \rightarrow NO_2 + O_2 \tag{4}$$

$$NO_2 + O \rightarrow NO + O_2 \tag{5}$$

$$\text{Net: } O_3 + O \rightarrow O_2 + O_2 \tag{6}$$

The long lifetime of the NO_y constituents allows the influence on ozone to last for a number of months to years past the event. Long-term effects due to solar protons with durations of (a) about 2 months have been shown in measured NO_x ($NO + NO_2$) and ozone after the July 2000 SPE (Randall et al. 2001); and (b) about 5 months have been shown in measured NO_2 and ozone after the October 1989 SPEs (Jackman et al. 1995). Figure 2 shows the model predicted temporal behavior of profile ozone (lower plot) and NO_y (upper plot) for the polarmost northern hemisphere (NH) area (70°–90°N) for the time period 1963–2010. NO_y shows enhancements between 1% and about 10% in the lower stratosphere (below 10 hPa) for particular years. These very large SPEs in (a) August 1972; (b) August–September–October 1989; and (c) July and November 2000, September and November 2001, October 2003, and January and September 2005 cause the NO_y increases in years (a) 1972–1973; (b) 1989–1993; and (c) 2000–2006, respectively.

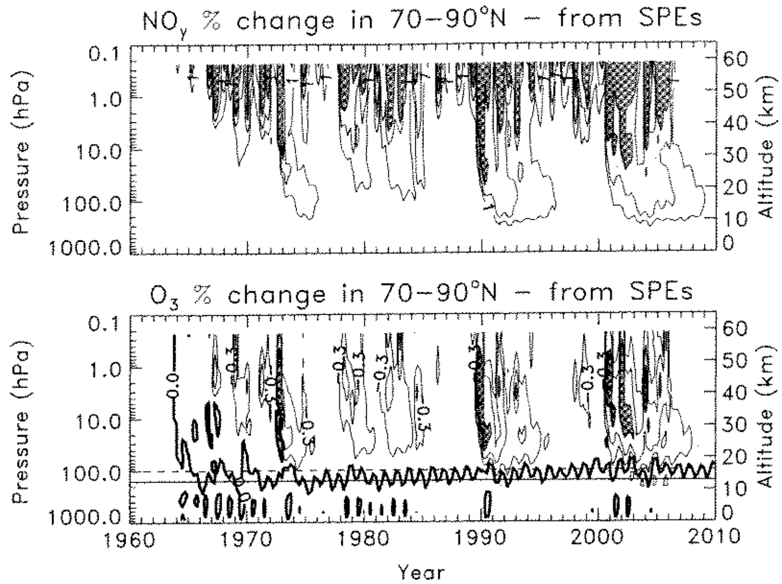

Fig. 2 Model computed percentage changes in NO_y and O_3 for the polar northern hemisphere area (70–90°N) for 1963–2010 resulting from SPEs in 1963–2005. Contour levels for NO_y (top plot) are +1%, +3%, and +10%. The "light gray" and "dark gray" highlighted areas for NO_y indicate increases from 3% to 10% and >10%, respectively. Contour levels for O_3 (bottom plot) are −3%, −1%, −0.3%, 0%, and +0.3%. The "light gray" and "dark gray" highlighted areas for O_3 indicate decreases from 1% to 3% and >3%, respectively. These changes were computed by comparing the "perturbed" to the "base" simulation. Horizontal lines at 160 hPa (solid) and 100 hPa (dashed) are given in the O_3 plot for assessing the temporal change of the 0% contour.

The increased NO_y led to a northern polar stratospheric ozone depletion for extended periods. SPE-caused depletions greater than 3% are highlighted in "dark gray" in Fig. 2 (lower plot). The polar southern hemisphere (SH) shows similar behavior (Fig. 3), however, there are differences caused by the seasonal differences for the occurrence of the SPEs (e.g., see discussion in Jackman et al. 2005b). SPEs that occur in the late fall/winter time period experience a lower amount of sunlight thus the loss process for NO_y via

$$NO + h\nu\,(<191\,nm) \rightarrow N + O \tag{7}$$

followed by

$$N + NO \rightarrow N_2 + O \tag{8}$$

is minimal. The vertical winds are generally downward at this time of year and NO_y is transported to lower altitudes, where photochemical loss is even less. SPEs in October 1989, November 2000, November 2001, October 2003, and January 2005 were thus most important in the NH. Likewise, the SPEs in August 1972, August

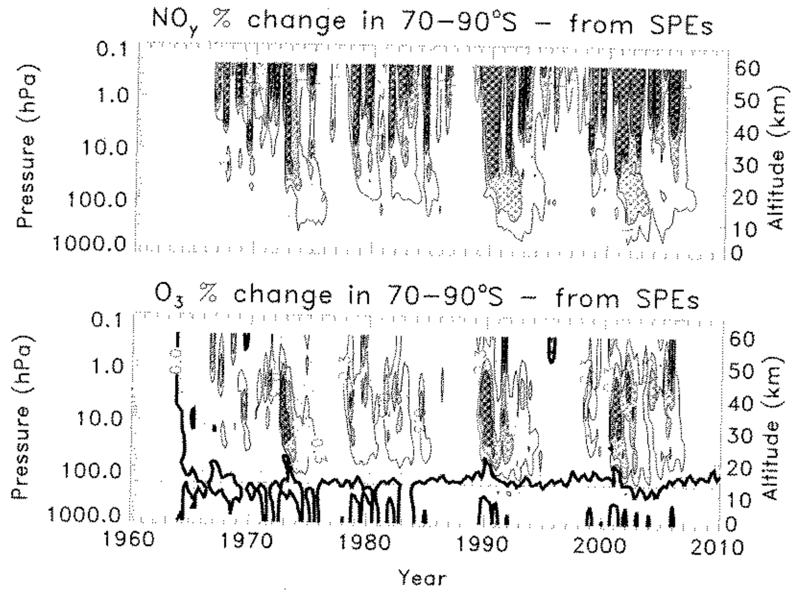

Fig. 3 Model computed percentage changes in NO_y (top) and O_3 (bottom) for the polar southern hemisphere area (70–90°S) for 1963–2010 resulting from SPEs in 1963–2005. Contour levels are the same as in Fig. 2. These changes were computed by comparing the "perturbed" to the "base" simulation. Horizontal lines at 160 hPa (solid) and 100 hPa (dashed) are given in the O_3 plot for assessing the temporal change of the 0% contour.

1989, and July 2000 were most important in the SH. Large SPEs, whether or not they occurred in late fall/winter, do cause perturbations in both hemispheres. For example, the October 1989 SPE was the largest in the past 40 years (see Table 1) and also caused a substantial impact in the SH (e.g., Jackman et al. 1995).

Enhanced levels of NO_y can also lead to ozone increases (Jackman et al. 2000). This is especially true in years of enhanced halogen loading. The ozone loss rate due to chlorine and bromine can be reduced through reactions such as

$$ClO + NO_2 + M \rightarrow ClONO_2 + M \tag{9}$$

and

$$BrO + NO_2 + M \rightarrow BrONO_2 + M \tag{10}$$

where chlorine and bromine reservoir constituents ($ClONO_2$ and $BrONO_2$) are produced at the expense of the ozone-reducing radicals (ClO and BrO).

Although such interference is relatively small in the NH and SH with computed ozone increases of just over +0.3% at most, the average altitude of the 0.0% contour line gradually rises upwards from about 160 hPa (~13 km) in 1980 to near 100 hPa

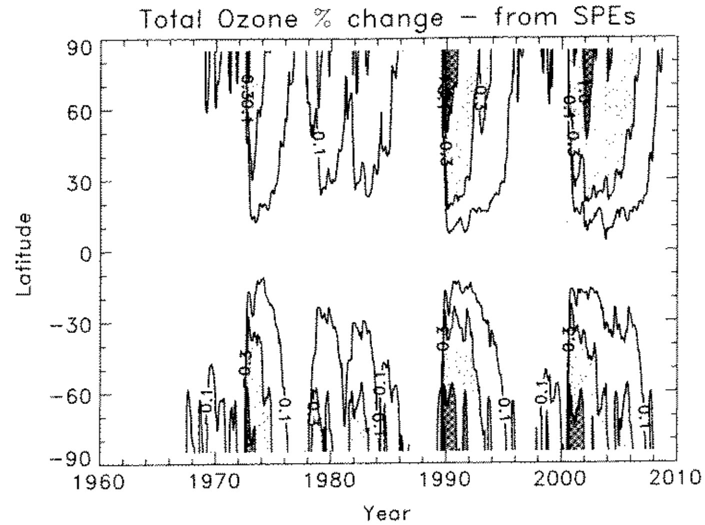

Fig. 4 Model computed percentage total ozone changes for 1963–2010 resulting from SPEs in 1963–2005. Contour intervals are –1%, –0.3%, and –0.1%. The "light gray" and "dark gray" highlighted areas indicate decreases from 0.3% to 1% and >1%, respectively. These changes were computed by comparing the "perturbed" to the "base" simulation.

(~16 km) in 2000 as the effective equivalent stratospheric chlorine increases from 1.8 to 3.2 ppbv over the same time period (see Fig. 1–23 of WMO 2003). There is significant seasonal variability (primarily driven by the annual solar change) and interannual variability (primarily associated with very large SPEs) in the 0.0% ozone contour line. Horizontal lines at 160 hPa (solid) and 100 hPa (dashed) are given in the bottom panels of Figs. 2 and 3 to aid in assessing the temporal change in the 0.0% contour line for ozone.

The impact on total ozone is shown in Fig. 4. Both hemispheres had extended periods of depleted ozone from 1–3% in 1972, 1989–1990, and 2000–2003. These changes are modest compared to the downward trends caused by halogen loading in the polar regions over the 1980–2000 time period [~3%/decade in the NH and ~4–9%/decade in the SH, WMO 2003], however, they need to be considered in understanding polar changes during particular years.

6 Conclusions

Several very large SPEs have occurred over the 43-year time period 1963–2005. These events, which occurred in 1972, 1989, 2000, 2001, and 2003, have led to significant polar enhancements in HO_x and NO_y. The HO_x enhancements led to short-term mesospheric and upper stratospheric ozone decreases, whereas the NO_y enhancements led to polar total ozone depletions of 1–3% lasting several months to years past the events. In more recent years, the NO_y enhancements were also found to lead to small ozone enhancements in the lowermost stratosphere because of interference with the ozone loss cycles for the chlorine and bromine constituents.

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